

Attorney Docket No. MSU 4.1-643
Appin. Serial No. 10/691,328
Declaration dated February 28, 2007



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Appln. No. : 10/691,328 Confirmation No. 1409
Applicants : Aaron L. Odom and James T. Ciszewski
Filed : October 22, 2003
Title : IMIDO-TETHERED CARBENES OF
MOLYBDENUM FOR RING-OPENING METATHESIS
POLYMERIZATION AND RING-CLOSING
METATHESIS
TC/A.U. : 1621
Examiner : Porfirio Nazario-Gonzalez
Docket No. : MSU 4.1-643
Customer No. : 21036

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DECLARATION UNDER 37 CFR 1.131/1.132

Sir:

We, Aaron L. Odom and James T. Ciszewski state as follows:

(1) We are the inventors of the above entitled invention;

2) In reference to the above entitled

application and the publication entitled Ciszewski et al., "Synthesis and structure of an imido-tethered Schrock carbene of molybdenum", Dalton Trans., pages 4226-4227, published on the web 09/25/2003 has been cited;

(3) The invention was made by us prior to September 25, 2003 at Michigan State University, East Lansing, Michigan. Enclosed is a copy of our Invention Disclosure filed with the University showing the conception and reduction to practice at least as early as February 4, 2003, well prior to the date of the publication in the Royal Society of Chemistry;

(4) That Baohan Xie is not an inventor and was included as an author only because of routine work in our laboratory after the date of the Invention Disclosure; and

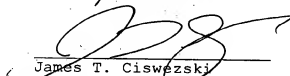
(5) That the undersigned declares further that all statements made herein of their own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States

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Code and that such willful false statements may jeopardize
the validity of the application or any patent issuing
thereon.


Aaron L. Odom

Date: 3/6/07


James T. Ciswewski

Date: 28 February 2007

Enclosure: Invention Disclosure

**MICHIGAN STATE
UNIVERSITY**

February 27, 2003

Ian C. McLeod, PC
Attorney and Counselor at Law
2190 Commons Parkway
Okemos, MI 48864

Fax: 347-4103

Re: MSU ID 03046F Imido-tethered Carbenes of Molybdenum for
Ring-Opening Metathesis Polymerization and Ring-Closing
Metathesis

Aaron L Odom

Chemistry

405 E. Miller Road, Lansing,
MI 48911

Robert Cichewicz

Horticulture/NFSTC

129 Felix St. Apt. 4

Dear Ian:

Please prepare and file a regular patent application on the subject technology upon approval of cost estimate. Background information via U.S. mail.

Sincerely,



Bradley T. Shaw
Licencing & Marketing Manager
Shawbr@msu.edu



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*Ian
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Dr. Odom
Brad*

1. Title of Invention:

Imido-tethered Carbenes of Molybdenum for Ring-Opening Metathesis Polymerization and Ring-Closing Metathesis

2. Name(s) of Inventor (s)

(List Primary Inventor First)

DEPARTMENT/COLLEGE

Aaron L. Odom Department of Chemistry
James T. Ciszewski Department of Chemistry

3. Sponsor (check all that apply)

☒ Yes ☐ No University funds, facilities or equipment used.

☒ Yes ☒ No Non-university sponsor

Sponsoring agency: Office of Naval Research

University research account number: 61-3475

☐ Yes ☒ No Other, describe...

☐ Yes ☒ No Was there a consulting relationship with sponsor?

4. Have any gifts (e.g., funds, equipment and/or supplies) been received that may be related to the invention?

If so, list:

5. Disclosure

☐ Yes ☒ No Invention has been disclosed to non-University personnel

If Yes, to whom?

Form of disclosure:

☐ Oral presentation ☐ Written format

☐ Yes ☐ No Was there a confidentiality agreement existing at time of disclosure?

Date disclosed:

☒ Yes ☐ No Disclosure, publication or presentation planned?

If Yes..

When: in 2-3 months

Where: communication to journal

To Whom: journal currently undecided

6. Date of Conception of the Invention:

E. List any key words to describe the invention.

See attached sheet

F. Include any other pertinent information relevant to the invention.

none

9. Please attach a nonproprietary description/abstract of the invention (one or two paragraphs).
see attached

10. All inventors should sign below; if necessary attach an additional signature sheet.

11. This Invention Disclosure should be reviewed and witnessed after preparation by one knowledgeable in the field of invention. The witness should sign below:

Disclosed to and understood by me,

Inventor's Signature

Mitch Smith
Typed/Printed Name

2/4/03
Date

Aaron L. Odom
Aaron L. Odom

James T. Ciszewski
James T. Ciszewski

4 Feb 2003

October, 2000

7. Have any prototypes, samples, etc., been made?

☐ Yes ☒ No

If so, when? several variations prepared in last few months

8. Use additional sheets to respond to the following and attach to this cover sheet.

A. Describe in general terms the objectives of the invention.

See attached sheets

B. List the advantages of this invention over previous approaches. What is the "old" method? What specific features of this invention make it more advantageous than the previous approaches? What markets could use this invention?

See attached sheets

C. Describe in detail the steps that need to be taken to implement or construct the invention (attach sketch if applicable). Are there alternate methods for implementation and/or construction?

See attached sheets

D. What is the stage of development of the invention now? Describe plans for continued future research and/or study on the invention.

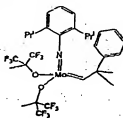
See attached sheets

A. Describe in general terms the objectives of the invention.

The goal of the invention is to develop a catalyst for ring opening cyclooligomerization of olefins. In addition, the catalyst design should provide for greater overall stability of catalysts, e.g. Schrock's catalyst, for ring closing metathesis of olefins.

B. List the advantages of this invention over previous approaches. What is the "old" method? What specific features of this invention make it more advantageous than the previous approaches? What markets could use this invention?

The current technologies for ring closing metathesis utilize ruthenium carbenes (Grubbs' catalyst, Caltech) or molybdenum carbenes (Schrock's catalyst, MIT). The carbene is untethered, and at the end of the reaction a relatively unstable methyldiene, $M=CH_2$, complex is generated, which often undergoes rapid decomposition. With the invention disclosed here, the methyldiene would be trapped to give a more stable catalyst at the end of the reaction. This in conjunction with current technologies for attaching the complexes to polymer supports will help to bring the technique to the mainstream of industrial catalysis. Currently, solid-supported catalysts are not reusable. With the developments in this disclosure they will become reusable and economically viable.

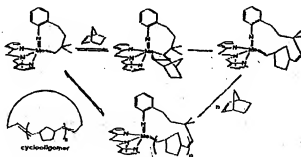


Schrock's Catalyst



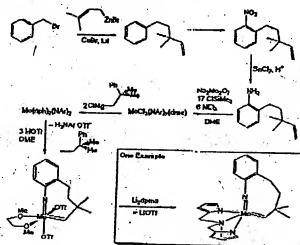
Grubbs' Catalyst

In addition, the catalysts offer new opportunities in materials and biological chemistry. The catalysts selectively polymerize cyclic olefins to form large macrocycles, which are currently quite difficult to prepare. A scheme describing the strategy and potential mechanism of cyclooligomerization is provided below.



The synthesis and an example of one of the active catalysts generated so far is given in the scheme

below. Many of the compounds in the imido ligand have not appeared in the literature. In addition, all of the metal complexes described in the scheme have not appeared in the literature. The reaction scheme culminates in the synthesis of a molybdenum pyrrolyl complex. We have generated several molybdenum catalysts from the bis(triflate) precursor shown. However, the pyrrolyl complex is quite stable and sufficiently reactive. Some future applications will entail having the ancillary ligand in that position attached to a polymer surface, chiral ancillary ligands, or both.



C. Describe in detail the steps that need to be taken to implement or construct the invention (attach sketch if applicable). Are there alternate methods for implementation and/or construction?

The invention is nearing a stage a full development. As mentioned above other ancillaries in place of the pyrrolyl ligand listed can be explored to increase functionality. Even though the catalyst will rapidly move towards maturity, the development of applications for the catalysts is in its infancy.

During development of our catalyst system, Grubbs' reported a system based on ruthenium for cyclooligomerization, which is the major competitor for the disclosed invention. In our hands, the molybdenum-based catalyst has proved much more reactive and generally applicable.

D. What is the stage of development of the invention now? Describe plans for continued future research and/or study on the invention.

We are beginning to explore applications of the catalyst in several different areas as we are completing full characterization/optimization of all the intermediates involved.

In ring-closing metathesis, the catalyst requires new ancillaries and testing of postulates regarding stability. Ring-closing metathesis is rapidly moving into the mainstream of organic synthesis in academia. In industry, some applications have begun to appear. The technology disclosed here will likely enhance the applicability of the molybdenum-based catalysts in industrial-scale processes. Grubbs' catalyst,

Schrock's catalyst, and modifications of both are commercially available from several sources. The modification disclosed here could rapidly be incorporated into existing product lines.

In ring opening cyclooligomerization (ROC), several untapped applications are going to be explored varying from new sensor technologies to mimics for Valinomycin-type antibiotics.

While the scheme above for ROC utilizes simple norbornene as the monomer, substituted norbornenes and a host of other cyclic olefins may be utilized. If, for example, ester substituted norbornenes are used a substituted ring would then be formed. The ester groups could be converted to acid chlorides using standard techniques. The acid chlorides could be reacted with diamines to form nylon-like linkages, which would polymerize the cyclooligomers. The character of the linkages could be altered from simple aliphatics that give novel materials to metal complexes. Since the metal would be part of the linkage, swelling of the polymer in the presence of various analytes will change the ligand field of the metal center. In other words, the color of the polymer will change on addition of various compounds. The extent of the change will be determined by the character of the polymer, which may be altered by changing the functional groups on the rings and in the linker.

Valinomycin is a member of a class of antibiotics consisting of a macrocyclic peptide ring. Like many peptide drugs, the compounds generally suffer from a lack of bioavailability resulting from degradation in the body. With the cyclooligomerization catalysts disclosed here, we could explore combinatorially a host of unnatural ring structures with various functional groups around the periphery that may act as mimics for members of this antibiotic class. With the unparalleled ease of macrocyclic ring synthesis enabled by this class of catalyst, we can rapidly explore combinatorial space for these mimics.

Other potential applications include additives for standard linear polymers that may prevent dewetting, which would be important for coatings and permanent data storage. The catalysts may also provide a means for the synthesis of cyclic polyacetylenes, which would be of potential academic and industrial interest.

E. List any key words to describe the invention.

cyclooligomer, olefin metathesis, imido, carbene, molybdenum, polymerization, pyrrolyl, macrocycle

9. Please attach a nonproprietary description/abstract of the invention (one or two paragraphs).

Through the use of an imido-tethered design on molybdenum carbenes, a catalyst motif for ring-closing metathesis that avoids potential decomposition pathways due to methylidyne formation and catalysts appropriate for ring-opening cyclooligomerization (ROC) of olefins is disclosed. The catalyst design incorporates a tethered imido ancillary ligand into the alkylidene functionality with no β -hydrogens and a linker of sufficient length to avoid potential problems with ring strain. Through the use of this tether, the local concentration near the metal active site of both ends of the oligomer is kept high during polymerization. As ancillary ligands, chelating pyrrolyl-based ligands and alkoxides have been utilized.

The cyclooligomers generated in this fashion are proposed as building blocks for a number of highly specialized polymers. The cyclooligomers described here are of norbornylene derivatives. Many of these monomers are commercially available and relatively inexpensive. The functionalized macrocycles may be polymerized using various crosslinkers. By varying the type of crosslinker and the functionality on the cyclooligomer, a number of applications follow naturally. The polymers are proposed for applications in purification of water and air, ion exchange, sensing of specific analytes through molecular recognition in combination with metallated cross-linkers, chemoselective supports for catalysis, synthesis of mimics for vancomycin-type antibiotics, as additives to prevent polymer dewetting, and as a potential means of accessing cyclic derivatives of polyacetylenes.

Synthesis and structure of an imido-tethered Schrock carbene of molybdenum†

James T. Ciszewski, Baohan Xie, Changsheng Cao and Aaron L. Odom*

Department of Chemistry, Michigan State University, East Lansing, MI, USA.

E-mail: odom@cem.msu.edu

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An 8-membered molybdenum azametallacycle, where a Mo–C bond and a Mo–N bond are linked, incorporating two triflate co-ligands has been synthesized and structurally characterized.

Metal–carbon double bonds where the metal is in a moderate to high oxidation state, Schrock carbenes or alkylidenes, seem to have an ever-increasing importance in small molecule and polymer chemistry due to their activity in double-bond metathesis.^{1,2} Since the original discovery of high oxidation state carbenes by Schrock in 1973,³ a plethora of carbene complexes in moderate to high oxidation states have been prepared across much of the periodic table.⁴ Some of the most active double-bond metathesis catalysts are the molybdenum(vi) imido carbene complexes, which are prepared by addition of alkoxides to bis(triflate) precursors (Equation 1). The triflate ligands may be replaced by a variety of different alkoxides,⁵ including chiral ancillaries⁶ for use in enantioselective ring-closing metathesis and solid-supported alkoxides.⁷

Fürstner and co-workers provided the initial reports on tethering the reactive ruthenium carbene functional group to an *N*-heterocyclic carbene ancillary ligand (Fig. 1).⁸ These carbenes were investigated for ring-closing metathesis as catalysts capable of regenerating the original species on depletion of substrate. More recently, Grubbs and coworkers have examined tethered ruthenium carbenes for selective cyclopolymerization of olefins by ring-opening metathesis polymerization.⁹ The tether retains a high local concentration of both termini of the growing polymer near the metal, enabling a final macrocyclic ring-closing metathesis event.



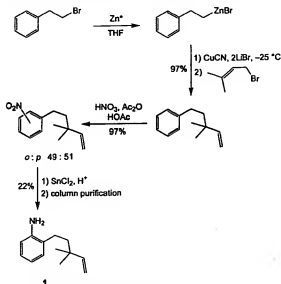
Fig. 1 Ruthenium catalyst with carbene tethered to ancillary ligand.

In a similar vein, we have been investigating the chemistry of tethered molybdenum carbenes, where the active carbene centre is attached through an aliphatic chain to an ancillary ligand on molybdenum. Like its ruthenium counterparts, it is hoped that

tethered catalysts would provide greater catalyst reusability by trapping less stable methylenide intermediates once substrates are consumed. In addition, cyclo-oligomerization and -polymerization products with numerous possible applications may be accessible utilizing these new molybdenum complexes.

To generate a stable tethered carbene and provide a bis(triflate) precursor useful for a variety of purposes, it was desirable to use the imido substituent as the ancillary ligand of attachment. However, relatively few complexes are known where two molybdenum metal–ligand multiple bonds are tethered to form a metallacycle. A few Group-6 mutually tethered di-carbenes have been reported, usually prepared through oligomerization of alkynes by reduced complexes.¹⁰ Tethered Group-6 bis(imido), *ansa*-di(organimido), complexes have been prepared as analogs of *ansa*-metallocenes of the Group-4 elements and studied for Zeigler–Natta polymerization activity.¹¹ The typical ring sizes used in the generation of tethered organimidos are 7–8 membered metallacycles. Consequently, we sought a ring size in this regime.

The successful strategy for the synthesis of the tethered carbene involved attachment to the *ortho*-position of an arylimido and generating an 8-membered azametallacycle. The synthesis of the aniline derivative is outlined in Scheme 1. The synthesis begins with commercially available 2-bromoethylbenzene, which is converted to the alkyl zinc by reaction with Reike zinc. Copper-mediated addition to 3,3-dimethylallyl bromide¹² occurs with the desired *S_N2'* to *S_N2* addition in a ratio of ~9:1 in 97% combined yield of isomers.¹³ The undesired isomer was not removed at this point, and the mixture was nitrated under standard conditions¹⁴ in high yield. The nitration occurs to generate a mixture of *ortho* and *para* isomers in a near equimolar ratio. Again, the mixture was carried forward

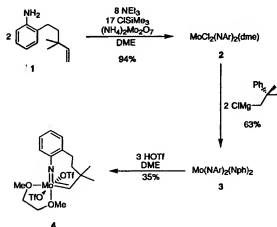


Scheme 1 Synthesis of 2-(3,3-dimethylpent-4-enyl)aniline (1).

† Electronic supplementary information (ESI) available: Synthetic details for the generation of 2-(3,3-dimethylpent-4-enyl)aniline (1) and the tethered molybdenum carbene 4. Tables for the X-ray diffraction study on 4. See <http://www.rsc.org/suppdata/dt/b311320p>

without separation of isomers. Reduction of the nitro group¹⁵ provided a mixture of aniline derivatives that were readily separable by column chromatography. The final step gave 22% yield of the desired *ortho*-substituted aniline **1** based on the crude mixture of nitro isomers.

The synthetic protocol for molybdenum carbene synthesis is a modification of the literature procedure (Scheme 2).² Reaction of **1** (H_2NAr) with NEt_3 , CisMoO_3 , and $(\text{NH}_4)_2\text{Mo}_2\text{O}_7$ in DME provided 94% yield of $\text{Mo}(\text{NAr})_2(\text{Cl})_2(\text{DME})$ (**2**). Alkylation of **2** with 2 equiv. of neophylmagnesium chloride (NphMgCl) afforded 64% yield of $\text{Mo}(\text{Nph})_2(\text{NAr})_2$ (**3**). Addition of 3 equiv. of triflic acid in DME removes one imido ligand as the anilinium triflate and presumably generates an intermediate neophylidene bis(triflate), which was not observed. Instead, the molybdenum carbene undergoes rapid cyclization with the alkene pendant to the arylimido producing the desired tethered carbene in 35% isolated yield. Examination of the bis(triflate) by multinuclear NMR in d_6 -THF suggested 3 isomers were present in fluid solution, a complication also observed in untethered systems.⁴



Scheme 2 Synthesis of imido tethered carbene.

Bis(triflate) **4** exhibits low solubility in many common solvents. However, crystals sufficient for a preliminary X-ray diffraction study were obtained.[†] The isomer observed is line drawn in Scheme 2, and a structural representation from the diffraction study is shown in Fig. 2. When the structure of

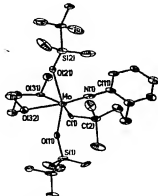


Fig. 2 Structure of **4** as found by X-ray diffraction. Selected bond distances (Å) and angles (°): $\text{Mo}(\text{O})-\text{Cl}(1)$ 1.827(19), $\text{Mo}(\text{O})-\text{N}(1)$ 1.723(16), $\text{Mo}(\text{O})-\text{Cl}(1)-\text{Cl}(2)$ 139.7(12), $\text{Mo}(\text{O})-\text{N}(1)-\text{Cl}(1)$ 173.3(14), $\text{N}(1)-\text{Mo}-\text{Cl}(1)$ 101.4(7).

4 is compared to that reported for $\text{Mo}(\text{OTf})_2(\text{N}-2,6\text{-diPr-C}_6\text{H}_3\text{(neopentylidene)(DME)})$,¹⁶ no ring strain is apparent in the metallacycle as judged by comparison of bond distances and angles.

As would be expected, the tethered carbene **4** is an active catalyst for the ROMP of norbornylene. The materials generated in those polymerizations are currently under scrutiny. Various substitution reactions on the triflate groups are being investigated as a means of synthesizing more functional metathesis catalysts.

The authors thank the Office of Naval Research, Department of Energy—Defense Programs, Petroleum Research Fund, and Michigan State University for Financial Support. JTC thanks MSU for Brubaker and Dye Fellowships.

Notes and references

[†] The crystals isolated were relatively weak diffractors, but a satisfactory structure was obtained. Crystal data for $\text{C}_{20}\text{H}_{24}\text{F}_6\text{MoNO}_8$: $M = 657.45$, monoclinic, $a = 12.669(3)$, $b = 15.745(5)$, $c = 12.809(4)$ Å, $\beta = 92.648(6)^\circ$, $V = 2552.3(12)$ Å³, $P2(1)1$, $Z = 4$, $\rho(\text{Mo-K}\alpha) = 0.764$, $T = 176(2)$ K, 11563 reflections, 3698 unique ($R_{\text{int}} = 0.2814$). The final $R(F) = 0.2494$ and $R(F) = 0.1088$ for reflections $I > 2\sigma$. CCDC reference number 217504. See <http://www.rsc.org/suppldata/b3/b311320.pdf> for crystallographic data in CIF or other electronic format.

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